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Superdomain Dynamics in Ferroelectric-Ferroelastic Films: Switching, Jamming, and Relaxation

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Abstract:

Recent experimental work shows that ferroelectric switching can occur in large jumps in which ferroelastic superdomains switch together, rather than having the numerous smaller ferroelectric domains switch within them. In this sense the superdomains play a role analogous to that of Abrikosov vortices in thin superconducting films under the Kosterlitz-Thouless framework, which control the dynamics more than individual Cooper pairs within them do. Here we examine the dynamics of ferroelastic superdomains in ferroelastic ferroelectrics and their role in switching devices such as memories. Jamming of ferroelectric domains in thin films has revealed an unexpected time dependence of $\tau^{-1/4}$ at long times (hours), but it is difficult to discriminate between power-law and exponential relaxation. Other aspects of that work, including spatial period doubling of domains, led to a description of ferroelastic domains as nonlinear processes in a viscoelastic medium which produce folding and metastable kinetically limited states. This $1/4$ exponent is a surprising agreement with the well-known value of $1/4$ for coarsening dynamics in viscoelastic media. We try to establish a link between these two processes, hitherto considered unrelated, and with superdomains and domain bundles. We note also that high- T_c superconductors share many of the ferroelastic domain properties discussed here and that several new solar cell materials and metal-insulator transition systems are ferroelastic.

I. Introduction:

Domains in ferroelectrics and ferromagnets have been studied for nearly a century now, but several ideas have been emphasized only very recently: These include nonlinear behavior, such as wrinkling, folding, coarsening and jamming.[1]

In a wide area of physics, ranging from polymer science to geology, the question arises of whether data for a position $x(t)$, such as a domain wall, can best be described by power-law relaxation

$$x(t) = A (t/\tau)^{-n} \quad (1.)$$

which is sometimes historically referred to as the Nutting Equation.[2],

or to an exponential decay

$$x(t) = A \exp (-t/\tau) \quad (2.)$$

or a stretched exponential decay

$$x(t) = B [\exp (-t/\tau)]^\beta, \quad (3.)$$

which is sometimes referred to as Kohlrausch-Williams-Watts relaxation,[3] in which the early work of Kohlrausch was later shown by Williams and Watts to be applicable to polymers.

The conclusion of the present review is that these questions are now being asked with regard to relaxation of ferroelectric and ferroelastic domain walls in thin films, and we suggest that present data are yet insufficient to make unambiguous discriminations. Moreover, as shown in the references discussed below, especially the work of Kubát and Rigdahl and of Weron, that for short times the stretched exponential description may generally prevail, whereas for long times, a simple power-law is asymptotically obtained.

There may be a connection between these relaxation studies in domains and coarsening. Coarsening is a phenomenon in which grains in ceramics or metals enlarge with time. It is known to exhibit a temporal dependence given typically by $t^{-1/4}$, [2-4] especially at long times (10-1000 s). [5,6] Nonlinear creep of polarization $P(t)$ is also found to give an exponent of $1/4$ in PZT at low stress, [7] where the authors refer to this as the Andrade power law. [8-10] The idea of coarsening has not often previously been extended to wrinkles and domains, rather than grains, but except for very short times the dynamics may be similar in the two cases: [11,12] Put simply, a different kind of coarsening creates superdomain “bundles” which display both ferroelastic and ferroelectric properties [13]. In [11] X. Chen and [12] S. Chen et al. describe the coarsening of wrinkles in viscoelastic layers and of jamming in magnetic domains; and in [14-17] Salje et al. and Zvelindovsky et al. describe coarsening in multidomain systems. They also show that shearing slows down coarsening of structures at later stages of phase separation, trapping the system in kinetically driven non-equilibrium metastable states. We note that Salje uses the term “coarsening” to refer to the broadening of crossed ferroelastic domains in a fine “tweed” structure to a coarser “tartan” structure. [15]

Salje has emphasized that these tweed structures can form a domain glass with a nonergodic response to external forcing. Such domain glasses can contain polar nanoregions, which are better known to exist in relaxor materials. He points out that complex domain structures, including tweed, may be stabilized by defects and that tweed structures are polar, either via the flexoelectric effect or via bilinear coupling between the strain and local dipole moments. Recently, Salje et al. [16] reported the first experimental evidence for piezoelectricity of a tweed structure in LaAlO_3 , where the uniform parent structure is centrosymmetric and shows no bulk polarity.

Both Chen and Zvelindovsky et al. found [12-18] power-law dependences with exponents very near $1/4$, as does Sinha [14]. There is a good earlier study of this problem by Kohn and Otto [19] based upon surface diffusion. A slightly different temporal exponent of $-1/3$ is obtained in the earlier model(s) of Wagner [20] and of Lifshitz and Slyozov.[21] To put things into recent perspective, however, as recently as 2011 it has been determined [22] that “there is as yet no physical interpretation for the loss mechanism” describing relaxation of 90-degree walls in the most popular ferroelectric, lead zirconate-titanate (PZT). This agrees with the earlier opinion on exponential versus power-law relaxation [23] that “Current theories of solid state flow cannot provide an explanation of the $v\sigma$ -behavior observed.” Here v is activation volume and σ is stress. These authors suggest an empirical relationship, such that power-law relaxation occurs in both metals and polymers only when $v\sigma > 10 \text{ kT}$ (250 meV at ambient T), and exponential decay for smaller stresses. This behavior may be compared with the limit in magnetic domain relaxation mentioned below that depends upon the ratio of dipolar to exchange energies.

Jamming and coarsening of domains are not equivalent phenomena, since in jamming the average size of the domains need not change; that is, very small domains are not necessarily annihilated to be incorporated into larger domains, although average domain areas may increase with time to minimize perimeters. Conversely, coarsening can occur for systems such as solid grains in a liquid or glassy matrix in which the grain boundaries rarely touch each other (Ostwald ripening [24]). Therefore it is useful to examine carefully over what range of time, temperature, stress, electric field or local electro-chemical ionic distribution the dynamics of domain coarsening and jamming are similar. Very recently the relationship between coarsening and jamming has been examined in the special case of foams.[25] Both coarsening and jamming arise to minimize the surface energy at the domain walls. In the case of coarsening, the domain wall area (A) increases, while specific surface energy (γ) does not change

$$\Delta G = \gamma \cdot \Delta A, \quad (4.a)$$

That is, coarsening takes place only when there is no change in the domain wall energy during the domain wall motion. In the more general case, the domain wall energy itself can also vary

$$\Delta G = \Delta(\gamma \cdot A), \quad (4.b)$$

e.g. due to change in defects or pinning sites that arise due to ferroelastic domain wall reorganization.[26]

A second question to consider is whether coarsening in multiferroics leads to superdomains or “bundles.” Fig.1 (left) shows a typical ferroelectric with 180-degree in-plane domains in which polarization $\langle P \rangle$ averages to zero, but strain $\langle S \rangle$ is non-zero (differing significantly along or perpendicular to the polarization in each layer). Fig.1 (right) shows the converse effect involving in-plane 90-degree domains, in which $\langle S \rangle$ averages to zero, but $\langle P \rangle$ is nonzero. In very early years (1955) it was known that some sort of superdomain structures were formed in barium titanate; Fig. 2 illustrates [27] such a case. In many ferroelectric-ferroelastics, superdomains will form in which $\langle P \rangle$ and $\langle S \rangle$ both average to zero over mesoscopic distances, to minimize depolarization and strain energies, Fig. 3.[28-30] In other systems, especially smectics, it has been shown that nano-ferroelectric domains constrained inside such ferroelastic domains can be normal to the ferroelastic walls or tilted (as in [28]), with a possibility of a phase change from one to the other in between the ferroelastic walls.[29, 30]

We shall term these arrays “superdomains,” although some authors term them “bundles.”[31] The simplest of these structures are closure domains in which typically four structures of the type illustrated in Fig. 1 are arranged clockwise or counterclockwise in a closed group. These have been studied in some detail previously. [13, 32-34] In particular, we know that these arrays can be moved via an electron beam in an electron microscope,[35] and that they exhibit creep.[36,37]

It is often found that ferroelectric nano-domains are nested inside larger ferroelastic domains such that the polarization (and hence depolarization energy) averages mesoscopically to zero; but the opposite case, of ferroelastic domains nested inside larger ferroelectric domains seems rare. (A good exception however, is [38,39]

$\text{Sr}_x\text{Ba}_{1-x}\text{Nb}_2\text{O}_6$ with $0.60 < x < 0.75$ with nano-ferroelastic domains clustered around the fourfold channels in this tungsten bronze, inside 300-nm-diameter ferroelectric domains; and a more recent exception is the spinel GaV_4S_8 , where skyrmion-like ferroelastic domains of diameter 20 nm are found inside ferroelectric domains an order of magnitude larger.[40]) One might ask why, that is, why does the strain energy not often also average to zero in the same way? We believe that the answer is that strain is never screened whereas charge and polarization are for the relevant length scale. In this respect it is probably important that GaV_4S_8 is a semiconductor, or more precisely a Mott insulator,[41] so that screening differs in it compared with a typical more insulating oxide ferroelectric, and that the ferroelectric transition is order-disorder and not displacive.

In addition to systems in which ferroelastic domains are all larger than the ferroelectric domains, or are all smaller, there are some materials such as WO_3 that exhibit a whole hierarchy of ferroelastic length scales.[42] In that crystal there are two different coexisting domain widths, thin around pseudocubic $\langle 100 \rangle$ axes and thick around $\langle 110 \rangle$, neither of which satisfies the Landau-Lifshitz-Kittel Law for thickness dependence on the film thickness to the power 0.5: The thin domains vary to the power 0.6, and the thick ones to the power 0.4 (so that at asymptotically thick values, they become equal!).

Parenthetically we note that these domain walls can be either charged or neutral. Janovec showed forty years ago[43] that at the ferroelectric-paraelectric phase transitions, charged walls usually arise where there had been mirror planes and uncharged walls, where there had been twofold axes. In addition to these two kinds of ferroelastic walls (mirror plane and twofold axis), more recently it has been shown that ferroelastic fourfold axes also exist.[44]

These ferroelastic bundles switch as a block in some ferroelectrics,[45,46] resulting in hysteresis loops with very large steps. We emphasize this aspect of superdomains, because it will control ferroelectric switching in many thin-film devices, because most ferroelectric memory materials are also ferroelastic. In these superdomains, the interactions among the ferroelastic domains and domain walls control the switching dynamics, not the behavior of smaller ferroelectric domains inside each large ferroelastic cluster or bundle. This behavior is similar to the dominance of vortex-antivortex interactions over the Cooper-pair interactions in Type II superconducting confined geometries as described by the Kosterlitz-Thouless model.[47] Very recently it has been pointed out that surface creases in elastic films are analogous to Kosterlitz-Thouless instabilities.[48] Other authors have shown [49, 50] that Kosterlitz-Thouless melting within n -vertex Potts model descriptions requires vertices with $n > 4$ and does not arise from the threefold and fourfold vertices considered by Srolovitz and Scott[51] and characteristic of BaTiO_3 , PbTiO_3 , and PZT films considered here. We should note that our analysis is supported by a recent work by Sigov who suggested that defects in ferroelectrics can be correlated and comply with the Kosterlitz-Thouless model. [52] In the Kosterlitz-Thouless framework, the energy of system is determined by the vortex-antivortex energy binding, ρ . This energy represents also the superfluidic stiffness, i.e. the viscosity or vorticity of the system. In analogy, the ferroelastic-ferroelastic interactions in bundle domains also represent the viscosity of the system. We suggest that it is this rheological characteristic of the ferroelastic domain network in superdomains that gives rise to doubling, tripling and wrinkling.

A simple way to pose this question is to consider the time evolution of such bundles as a ferroic film is cooled below its Curie temperature, T_c . Do these extended arrays form to minimize strain and depolarization energy in thermal equilibrium? Are they non-equilibrium structures that are kinetically limited?

A series of studies of ferroelastic domain walls has been published by Salje, of which recent examples are given in [53-55]. These generally describe the kinetics via stretched exponentials rather than power laws. They do however introduce nonlinearities in the context of avalanches. Moreover, they show [56] that the jamming relaxation follows a power law at low temperatures but an exponential at higher temperatures. A similar conclusion was reached by Metzler and Klafter, who obtain stretched exponentials and power laws as the two asymptotical limits of a Mittag-Leffler equation.[57, 58] A good study of the interaction of ferromagnetic, ferroelectric, and ferroelastic domains has been given recently by Van de Wiele et al.[59]

It is more common to treat relaxation data for dielectrics to a stretched exponential, rather than a power law. However, Weron shows that the stretched exponential is merely a special limiting approximation to a more general power law.[60-62] She points out that the exponential arises when one neglects the distribution of waiting times for the relaxing dipoles and when one treats each relaxing dipole as independent, as contrasted with cluster behavior. The problem with many switching models (e.g., Avrami and nucleation-frustrated) is that they assume all relaxation starts at the same time $t=0$; that is, there is no “waiting time.” In reality several different mechanisms can contribute to such waiting time, including back-switching processes; the net result is that most of the relaxation process begins not at $t=0$ but at a finite delay time τ_0 . At best this introduces another fitting parameter to the data, and unfortunately this parameter τ_0 will be highly correlated with the exponent used in the least squares fit.

Many researchers have shown that these powers are fractional; and it is interesting to note that power-law relaxations in solids date back to the early 18th Century.[63] For magnetic domains the relaxation can follow either an exponential decay or a power-law decay, depending upon the ratio of dipolar to exchange interactions.[64]

More recently the non-Ising qualities of ferroelectric domain walls have been analyzed, and it would appear that some of the mysterious losses and relaxation may involve flexoelectricity, which is always present, independent of crystal symmetry, and estimated in PZT or BaTiO₃ as about 4% of the total polarization of the domain walls.[65,66]

Finally, readers are referred to an excellent treatise on ferroic domains by Jill Guyonnet for a discussion of these problems.[67]

II. Domain spatial period doubling and tripling:

Ref. [1] illustrates period doubling in ferroelastic domains in samples of lead zirconate-titanate iron-tantalate that have been subject to focused-ion beam cutting. There are two

questions that might be asked regarding those data: First, does it occur in other ferroelectric/ferroelastics? And second, does it require FIB processing to provide sufficiently nonlinear stresses? Fig. 4 answers these questions by showing domain period-doubling (top of figure) in PbTiO_3 films (on KTaO_3 substrates) that have not been subject to FIB. These are 22 nm thick. The thinner domains are about 12 nm wide, and the thicker, about 25 nm. These widths are not in accord with the Landau-Lifshitz-Kittel formula for a 22 nm thickness, given numerically by Ref. [68,69], which further supports their nonlinear and/or non-equilibrium origin.

Fig.5 illustrates a similar period doubling in non-epitaxial, thicker (60-nm) ceramic PZT films. The microscopic, atomistic dynamics are revealed in the dislocation line creation and annihilation shown in Fig.6. And in Fig. 7 we see a spatial tripling of domain width. Such a tripling was initially predicted in the nonlinear domain wall creep model of Metaxas et al.;[70] and Wang and Zhao showed[71] that it requires a higher strain rate than does doubling. The detailed model of Wang and Zhao merits a fuller discussion. Prof. Xuanhe Zhao and his postdoc Qiming Wang describe these regimes of wrinkling, folding, and period-doubling as separate thermodynamic phases, with true phase boundaries between them. These authors show how wrinkling in domains evolves into folding and subsequently to period doubling as a function of both stress and the ratio of film/substrate thickness, in a three-dimensional phase diagram. In general it is possible to go to frequency doubling without folding first (via increased strain), or to go from folding to period doubling directly via decreased film thickness. A qualitative feature of their model is that the thin film must have a larger modulus than the substrate; it forms a crust which must fold or wrinkle under compression to avoid delamination. Note that it has not been known in general how thick ferroelectric domains are (are they near-surface layers?) with a few exceptions[72] nor what their shear moduli are.

Not all models or systems exhibit spatial tripling: Brau et al.[73] show that a sequence of doubling, quadrupling, etc. can occur without tripling; such a sequence eventually leads to chaos. The same group [74] has shown that the period doubling is a continuous second-order phase transition and that it requires an up-down asymmetry; hence it is not present for their model in sandwich-structure thin films in which the lower substrate and top capping layer are identical.

III. Domain wall jamming relaxation: Power-laws or exponential decay?

Some sparse data on relaxation of domains in other ferroelastics exist (Fig.8). Fig. 8 illustrates data on the decay of surface potential around 90-degree walls in single-crystal BaTiO₃ [75-77]. The fits to exponential decay with $\tau = 30$ s and with a power law are both shown, illustrating the difficulty in unambiguous discrimination for typical published data. Note that for power laws with exponent $n \ll 1$, any data asymptotically approach a logarithmic (exponential) dependence.

These show that the relaxation time for 90-degree walls is ca. 100x faster than for 180-degree walls.[70] For 90-degree walls in PbZr_{0.3}Ti_{0.7}O₃ the relaxation time is typically 780 s, whereas for 180-degree walls it is ca. 10^5 s.[78] This compares with a jamming relaxation time of ca. 5000-7000 s in BaTiO₃,[32,33] suggesting that the jamming time (almost the same as 180-degree reversals) is not limited primarily by 90-degree wall relaxation. The diameter of the typical bundle or superdomain studied in [75] was 80 nm. This gives a ferroelastic area that typically contains ca. 100 ferroelectric nano-domains (see Fig.9, bottom diagram). The fact that the ferroelectric domains can be reoriented within a larger ferroelastic superdomain by application of electric field has been demonstrated by electron beam irradiation [34,35] with faceting and de-faceting observed.

There are three fundamentally different non-equilibrium patterns for domains: The one in the upper left panel of Fig.4 is usually termed a dislocation pattern; and the one in the lower left panel of Fig.9 is called a spiral pattern. These have quite different dynamical origins, discussed in theoretical detail by Hohenberg and Krekhov [79], who show that the dislocation pattern arises from their 'Type I.s stationary-periodic instability,' whereas the spiral pattern arises from their 'Type III.o oscillatory-uniform instability;' they also show a third 'target pattern' shaped like a bulls-eye, which was reported in ferroelectrics by Gruverman et al. and by Dawber et al. [80,81] and called a 'vortex pattern' or 'perimeter effect' but not reproduced here. In addition they show a 'domain boundary pattern' that can include 'herringbone' configurations. They comment that in this categorization the dislocation patterns have a mechanism similar to that in Rayleigh-Benard convection instabilities or Taylor-Couette instabilities with rotating inner cylinders, whereas the spiral pattern is related to the Belousov-Zhabotinsky reaction instability.

One should point out that the number of sides of the domains in jammed ferroelastic-ferroelectrics are not random but are influenced by the strong preference for vertices to be threefold in the materials considered.[51] This severely constrains available geometries. Hence, although jamming is sometimes described as chaotic, it is not geometrically random.

Finally, we note that the reverse of Ostwald ripening and jamming has been studied in magnets when the applied field is turned off: There is a decrease in size of domains and an increase in the number of domain walls as magnetization flops from out-of-plane to in-plane.[82]

IV. Switching of superdomains:

Fig.10 shows the TEM photos of a ferroelastic superdomain switching large amounts of charge.[83] This shows that ferroelastic superdomains move as large coherent blocks in ferroelectric switching [31,84-87] and hence that the switching dynamics are not dominated by reversal of nano-scale ferroelectric domains within these blocks, i.e. are not described by the Landau-Lifshitz-Kittel domain model.[88,89] the Landau-Lifshitz-Kittel model is also a linear response model and hence does not describe domain wall folding, which is a high-stress phenomenon[1,90]; such phenomena are also known in liquid crystals.[91]. Fig.11 illustrates this in a three-step ferroelectric switching in ultra-tetragonal PbTiO_3 . [46] The role of these ferroelastic blocks or superdomains is analogous to the role of Abrikosov vortices in thin films of Type II superconductors, where the vortex-antivortex pairs on the meso-scale dominates the dynamics of individual Cooper pairs. Ref [92] supports the conclusion [26,31] that ferroelastic “bundles of ferroelectric stripes constitute the macroscopic polarization.”

V. Conclusions:

Period doubling and tripling, wrinkling and folding in ferroelastic/ferroelectric domains support the early nonlinear models of Metaxas et al.[70] and of Wang and Zhao.[71] Folding models in elastic thin films seem applicable to thin ferroic domains. Some data exist suggesting the presence of power-law relaxation in the jamming of these domains, analogous to ripening, but existing data make it difficult to discriminate between exponential and power-law decay with powers <1 . This suggests a direction for further work. Other models that still require experimental testing include the predicted $d^{4/9}$ power dependence predicted for folding threshold stress upon film thickness d , [93] and the conjecture of Aharoni et al. that the wrinkle instability wavelength sets the scale for herringbone patterns.[94] In addition, the faceting of thin films confined to circular geometries[95,96] has also been described in terms of wrinkling.[97] we emphasize for readers that ferroelastic domains are of great importance not just in ferroelectrics, but in high-Tc superconductors, where so far they have always been found to satisfy the Landau-Lifshitz-Kittel law (square root dependence of domain width versus film thickness), from a few nm to ca. a micron.[98] Finally, in the context of period doubling and tripling, Roytburd et al. [98] have recently stressed in a comprehensive review on ferroelastics, that there may be situations in which two different kinds of domains are simultaneously present in quasi-equilibrium, due to boundary conditions, and these do not involve nonlinear

doubling or tripling. And Jang's laboratory has shown that the Landau-Lifshitz-Kittel law can fail in nanostructures due to surface layers.[99] Moreover, Noheda's group has shown that superdomains in $\text{PbSr}_x\text{Ti}_{1-x}\text{O}_3$ can dominate ferroelectric switching.[100]. These in-plane superdomains are strain-neutral, like those in the right-hand side of our Figure 1. It was found that fourfold vertex closure domains in this material are unstable, unlike those in BaTiO_3 [32,101] or PZT,[13] in agreement with the clock-model predictions of Srolovitz and Scott.[51] When treating in the clock model the domain walls, rather than treating the polarization itself,[52] it is clear that bundle-domain switching, which can be described as correlated motion of defects,[86] so that the ferroelastic switching indeed dominates the polarization switching, in analogy to the Kosterlitz-Thouless dominance in thin superconducting films..

Other ferroelectric/ferroelastic systems in which domain motion has been carefully studied include Bi_2WO_6 , in which the very high ferroelastic mobility is notable.[103]

Regarding power-law versus exponential relaxation, existing data leave this question as moot, suggesting that power laws dominate at long times and exponentials at short times.[21,56,104]

Ferroelectric memories are now rapidly replacing magnetic stripe cards for train and subway access, for cash points ("e-money"), employee identification cards, convenience store sales, luggage lockers, etc., where they are faster, cheaper, and contact-free [105] (proximity devices which can be read without removal from a wallet). It may be useful to recognize that the active material in use at present in these cards (from, for example, the SONY-family FeliCa or the Suica companies) use ferroelastic-ferroelectrics for which the switching mechanisms discussed above are operative. Other applications of multiferroic domains include the strong coupling of magnetic and ferroelectric walls through their mutual interaction with ferroelastic walls. Van der Wiele et al. show[106] that strong coupling of magnetic domain walls onto straight ferroelastic boundaries of a ferroelectric layer enables full and reversible electric-field control of magnetic domain wall motion.

In the context of the present paper the questions for commercial devices are how the ferroelastic properties might limit lateral cell size, switching speed, or reproducibility of switched charge. Perhaps more immediate concern is the role of ferroelastic domains on the performance (either enhancement or degradation of lifetimes) of new solar cell materials.[107] These may limit performance or provide degradation. Other aspects of ferroelastic domains of current device interest include their role in metal-insulator transitions in materials such as VO_2 . [108] It has been known for many years that ferroelastic domains increase toughening in PZT ceramics.[109] We encourage readers also to take advantage and implement the correlated switching in bundle domains for novel technologies, including those related to domain-wall engineering and in other metal-oxide applications.

Readers should keep in mind that many ferroelectrics of recent interest are not ferroelastic (e.g., YMnO_3). The criterion for ferroelasticity originally suggested by Toledano[110] was that it is necessary and sufficient that the crystal class change at the ferroelectric transition, such that rhombohedral-rhombohedral transitions (e.g., LiNbO_3) are not ferroelectric, nor are orthorhombic-orthorhombic (e.g., KTiOPO_4). However, this definition requires several exceptions, such as needing hexagonal and trigonal classes to be treated as a single super-class, and not discriminating between Laue Type I and Laue Type II tetragonal structures.[111] Therefore we propose a simpler definition that has no exceptions: Ferroelastic transitions are between phases having a different number of nonzero symmetry elements in the fourth-rank elasticity tensor. For example, Laue Type I and II crystals can both be the same tetragonal crystal class but differ in whether C_{16} is zero.[112]

VI. Future work:

Although the present review emphasizes net motion of domain walls, we note that oscillation of such walls is also a topic of current interest.[113-115] Chu et al.[116] have commented that the major contribution to the dielectric response is from the polarization fluctuations on the 90° -domain walls, which are more mobile than those inside the domains. The theory [113,114] predicts a gap energy in the acoustic phonon/soft-optic mode spectrum at a few GHz, which appears to have been found in ferroelectric trisarcosine calcium chloride.[115] Such low-frequency overdamped modes can arise from different physical mechanisms, including dynamics of incommensurate domain structures, and in a few cases have been shown[117] to be diffusive, with linewidth varying as q^2 , where q is the momentum transfer; this q^2 -dependence is a signature of hydrodynamic diffusion and in general supports the basic hydrodynamic model of domain wall motion under stress.[1,90] Another material of recent interest is HfO_2 , where [120] Depner et al. find ferroelastic domains narrower than 1 nm which are totally absent in bulk. This is in accord with the basic Landau-Lifshitz-Kittel idea that the characteristic length in the problem need not be a macroscopic thickness for all geometries.

In summary, to give a broader perspective on and support for the present work, although folding and bent curvature of domain walls in ferroelectric/ferroelastic crystals is a rather new topic, it should not be viewed as controversial or with skepticism: In the context of pure crystallography both folding and curvature are well known. For example, Ye et al. [121] comment in their TEM study of bismuth titanate, “domain walls (DWs) lie mainly on the (110) plane, but often fold to the (001) plane. ...The $P_{s(c)}$ - 180° DWs observed are irregularly curved.” And as early as 1957 Cameron reported “puddle” ferroelectric domains without straight walls in BaTiO_3 . [122]

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Figures and figure captions:

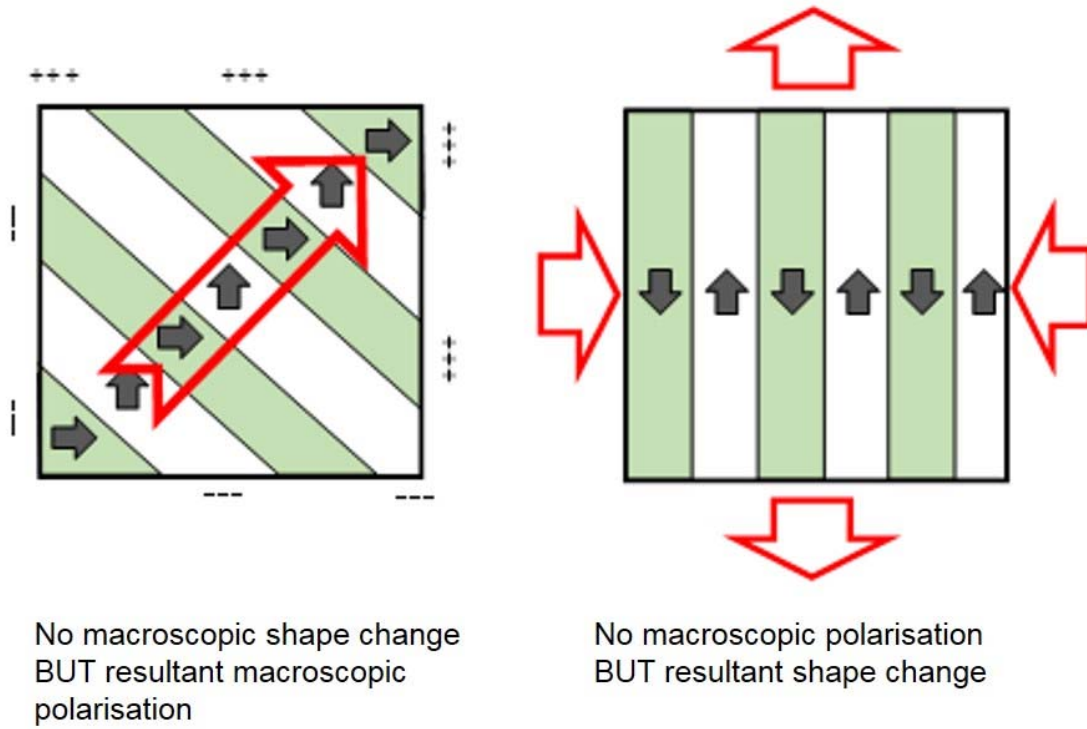


Figure 1. (a) Right-hand-side: 180-degree in-plane domains with average polarization $\langle P \rangle = 0$ but nonzero average strain $\langle S \rangle$ (indicated by large arrows); (b) Left-hand-side: 90-degree in-plane domains with average strain $\langle S \rangle = 0$ but nonzero average polarization $\langle P \rangle$, indicated by large arrow. This stress will produce domain wall motion. Both herringbone and closure domain structures produce $\langle S \rangle = \langle P \rangle = 0$.

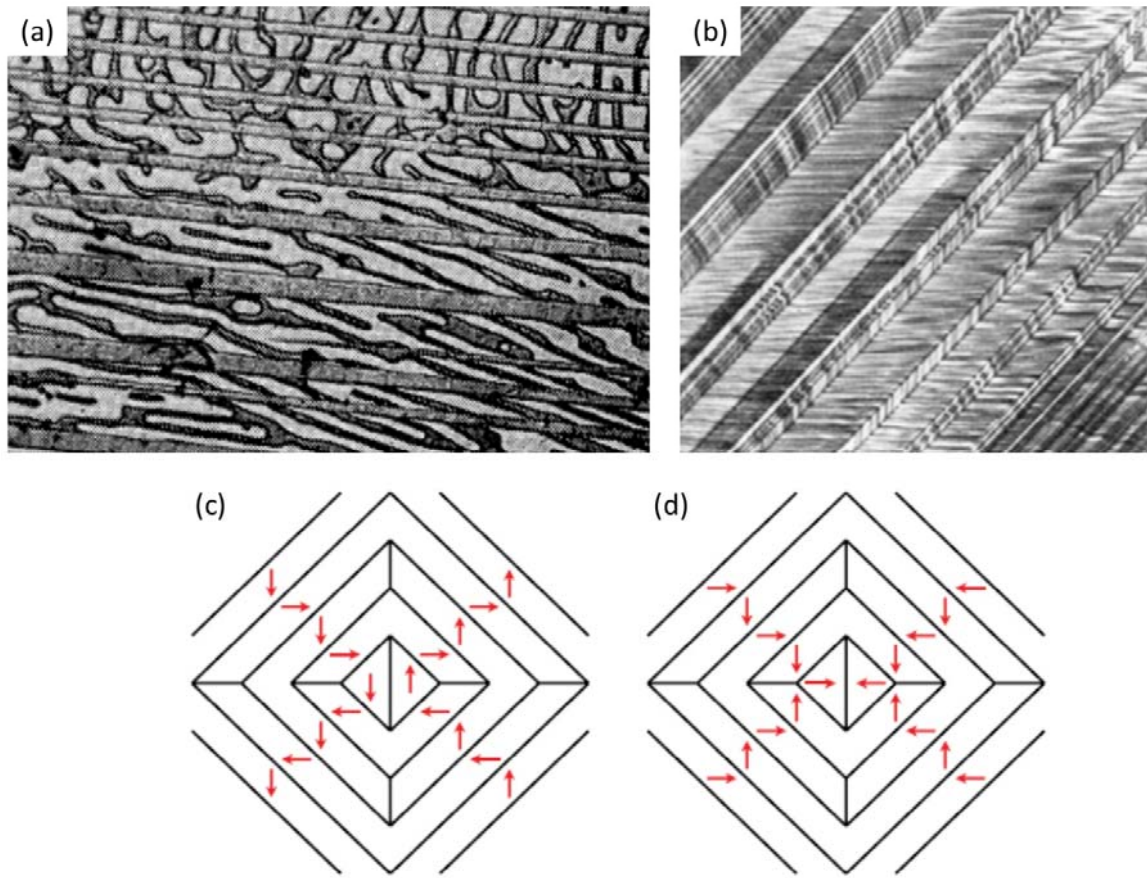


Fig. 2 (a): A real superdomain from early work on BaTiO₃: Stacks of flat ferroelastic 90-degree domains order with wavy 180-degree ferroelectric domains inside them. Note that some of the wavy domains are constrained by flat domain walls and some interpenetrate those walls. The 180-degree domain walls are very straight normal to the plane of the figure but hang in folds like drapes within the plan view. (b) Large ferroelastic domains each containing small ferroelectric nano-domains.[27] (reprinted with permission from J.A. Hooton, *Physical Review*, 98, 409, 1955.] Copyright 1955 by the American Physical Society. (c) Quadrupolar and (d) radial patterns of two possible arrangements of domains.

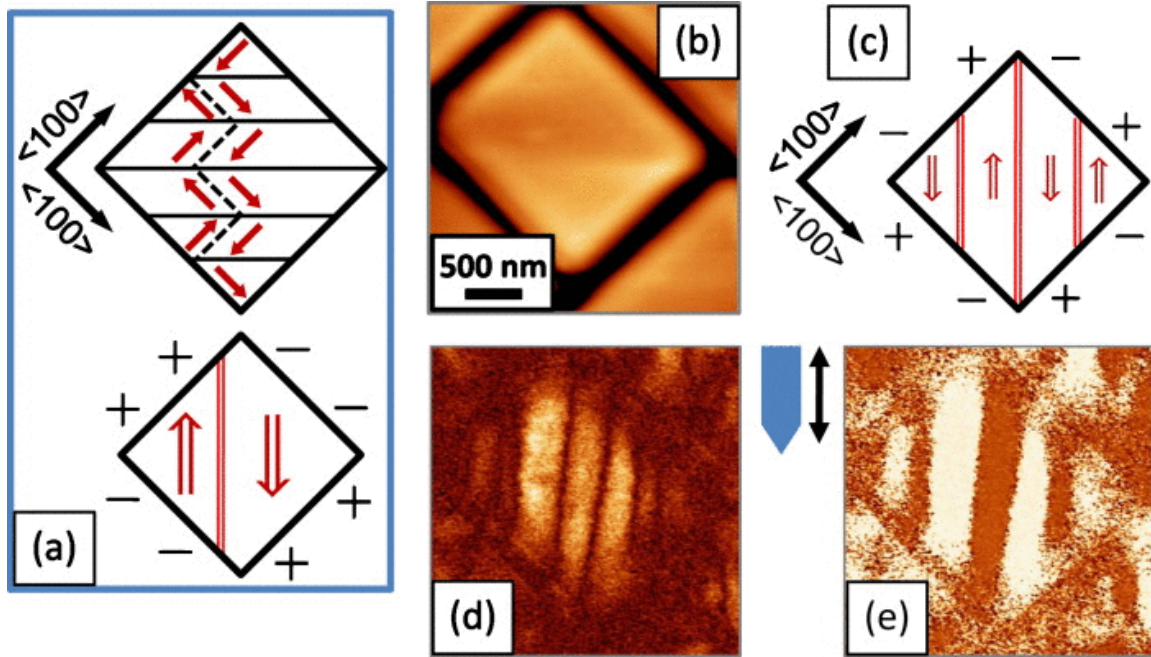


Fig. 3. A bundle or superdomain [84] that minimizes both strain energy and depolarization energy by forming a mesoscopic array with both $\langle P \rangle = \langle S \rangle = 0$: (a) Top: $\langle S \rangle = 0$ state in the core; bottom: $\langle P \rangle = 0$ state in the core. The top and bottom configurations do not have the same total energy. The bottom seems preferred in our materials, probably because charge is screened but strain is unscreened. (b) Experiment. (c) $\langle P \rangle = 0$ state at the core with spatial period halved in comparison with (a). (d) and (e) experimental data. Reprinted from [L. J. McGilly, J. M. Gregg, *Appl. Phys. Lett.* **98**, 132902 (2011)], with the permission of AIP Publishing

